Study of Ultrafast Photo-response of Organic Materials for All-Optical Computing

Synopsis

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Introduction

The study of ultrafast interaction of light with matter on ultrashort timescales, typically less than a picosecond, is important for both fundamental and applied science. It provides insights into the processes that occur in atoms and molecules and enables their optical characterization. Ultrafast interaction is achieved with lasers generating ultrashort pulses and transient absorption spectroscopy. Various nonlinear optical processes are involved in different materials at ultrashort time scales, which include, nonlinear absorption (NLA), multi-photon absorption, frequency conversion, self-phase and cross-phase modulation. They provide opportunity to control *'light with light'* that is necessary to realize ultrafast all-optical information processing (Genty et al., 2020; Sharma et al., 2020).

The bandwidth demand for high speed signal processing and optical communication systems is predicted to shift toward 100 to 400 Gbit/s in the next few years (Hommelhoff & Kling, 2015; Segatto et al., 2018). Due to fundamental speed limit of conventional semiconductor materials and devices, the existing optoelectronic and electronic devices and integrated circuits would not be able to respond beyond 100 Gbits/s (Flamini et al., 2018; Ishikawa, 2008; Mao et al., 2019; Wada, 2004). However, present-day light wave communication systems require higher processing speed and storage capacity, and need to overcome the limitation of speed and bit rate capacity due to optoelectronic conversion at the transmitter and receiver ends (Roy, 2011).

For the design of next generation communication systems, the ability to generate and control ultrashort light pulses has exciting implications, as they are necessary for future high bit rate optical communication systems. All-optical switching and signal processing with the high intensity ultrafast light pulses will offer tremendous bandwidth capacity for optical fiber transmission (Dudley et al., 2007; Idachaba et al., 2014; Wada, 2004).

Optical computing has emerged as a promising strategy by offering excellent features that include parallelism, high speed and temporal bandwidth, low signal dispersion, non-interactive behavior and high speed interaction with materials (Abdeldayem & Frazier, 2007; Fleischhauer, 2011; Kaku, 2000; Langford et al., 2011; Li et al., 2018; Touch et al., 2017).

To design all-optical ultrafast devices for communication and computing is a foremost challenge to achieve the next generation ultrafast speed and ultrahigh bandwidth information processing. This

requires extremely photosensitive and efficient nonlinear optical (NLO) material that exhibits fast response, strong nonlinearities to facilitate low-power switching, high photothermal stability, low optical loss, high efficiency, broad wavelength range, low cost, flexibility to alter the characteristics and also the ease to fabricate and integrate them into photonic systems (Abdeldayem & Frazier, 2007; Arivuoli, 2001; Belousova et al., 2007; Dudley et al., 2007; Imakita et al., 2012; Z. Lu et al., 2015; Ma et al., 2021; Messier et al., 2012; Miri & Alu, 2019; Ostroverkhova, 2018; Prasad & Reinhardt, 1990; Suresh et al., 2012; Wada, 2004).

The basic absorption processes in materials can be divided into linear and nonlinear absorption (NLA). NLA is an important NLO phenomenon defined as a nonlinear change (increase or decrease) in absorption with increase in intensity. Depending on the pump pulse intensity and absorption cross-section at the excitation wavelength, most molecules show NLA. This can be of two types: Saturable Absorption (SA) and Reverse Saturable Absorption (RSA).

In SA, when the absorption cross-section of the excited state is smaller in comparison with ground state, the transmission of the system increases on excitation of material, whereas, in RSA, when the absorption cross-section of the excited state is larger than that of the ground state, then the system becomes less transmissive when excited. These absorption characteristics are highly dependent on the wavelength, pulse intensity and excited-states lifetime. Ultrafast intense laser sources provide detailed knowledge of the absorption characteristics of a material. SA is invariably used in mode-locking and Q-switching, while RSA is useful for optical pulse processing, laser physics and optical limiting devices that protect sensitive optical components, including human eye, from laser-induced damage.

In recent years, intensity-dependent transitions from SA to RSA or RSA to SA have been extensively studied in nanosecond (ns), picosecond (ps) and femtosecond (fs) regime, in a variety of molecular configurations. SA has been recently studied for antimonene quantum dots (Wu et al., 2021), few layered germanium sulfide (Zhou et al., 2021), titanium trisulphide (Liu et al., 2020), polaritons (Raab et al., 2020) and RSA in vegetable assisted Au nanostructures (Emusani et al., 2020), natural pigment extracted from *coriandrum sativum* (Jeyaram & Geethakrishnan, 2020), SnS_2 (Diao et al., 2021), Aluminum doped zinc oxide thin films (Sreeja et al., 2021), ReS_2 (Mandal et al., 2021), PVA films doped by the novel natural dye extract from C. redflash leaves (Haripadmam et al., 2021), Penta(zinc

porphyrin)[60] fullerenes (Kulyk et al., 2020). Materials which show both SA and RSA together have been reported in molybdenum disulfide (C. Lu et al., 2020), $ZnCo_2O_4$ microflowers (Khan et al., 2020), silver nanoparticle (Maurya et al., 2019), Au- triangular nanoplates (Zhang et al., 2020). Interestingly, transition from SA to RSA has been reported recently in WS_2 (Karmakar et al., 2021), carmoisine dye (Abdullah et al., 2019), $MoTe_2$ nanofilms (Quan et al., 2018). These studies essentially focused on characterization of their NLO properties using Z-scan technique to understand the mechanism for NLA and refraction for potential application in optical limiting.

Compared with inorganic materials, various kinds of organic photoresponsive molecules that includes small organic molecules, metal complexes, polymers, supramolecules, cholesteric liquid crystals, organic dyes etc. have emerged as excellent materials for photonic applications, especially for all-optical switching, due to their unique advantages such as low power operation, high order nonlinear coefficients, easy tunability through molecular design, low manufacturing cost, ability to tailor properties for a specific purpose and the ability to integrate them with conventional devices.

Theoretical design of ultrafast all-optical logic gates and circuits have been reported for some organic materials that include phthalocyanines (Bhattacharya et al., 2019, 2021; Hussein & Mohammed, 2020; Ran et al., 2020; Roy & Yadav, 2011; Sebastian et al., 2020), Graphene, Graphene-oxide thin films (Bhamare et al., 2019; Meymand et al., 2020; Roy & Yadav, 2013; Shang et al., 2014; Tong et al., 2018; Yadav & Roy, 2015, 2014), Ruthenium-dioxolene (Cassano et al., 2005; Manjunatha et al., 2013; Tarnovsky et al., 2006), Bacteriorhodopsin (Cheng et al., 2012; Kovacs et al., 2019; Roy & Yadav, 2014; Smitienko et al., 2020), Cytochrome-c (Andrade et al., 2003; Boni et al., 2010; Yadav & Roy, 2016), Porphyrins (Biswas et al., 2020; Katturi et al., 2020; Rao et al., 2018; Wang et al., 2020), Chromophores (Arpa et al., 2020; Chen et al., 2020) and Chlorophyll (Boni et al., 2007; Ghanem et al., 2021; Jeyaram & Geethakrishnan, 2019; Roy et al., 2009).

Biomaterials have emerged as an important class of organic materials for photonic applications. Bacteriorhodopsin has emerged as one of the most prominent natural photodynamic protein which possess unique properties that include photochromic, photoelectronic and charge transport responses, high quantum yield, high two-photon cross-sections, long term photothermal and chemical stability, flexible for tailoring its properties, environment friendly and low production cost. All-optical switching

and computing has been reported for natural photoreceptor proteins using pump-probe configuration such as bacteriorhodopsin, *pharaonis* phoborhodopsin, cytochrome-c and chlorophyll-A (Roy et al., 2009; Sharma et al., 2005; Yadav & Roy, 2016) and also with BR protein coated microcavities (Roy et al., 2010, 2012), 2×2 port silicon micro-ring resonators coupled to optical fibers has also been reported (Chhipa et al., 2021; Ooi et al., 2017; Sethi & Roy, 2014a, 2014b). However, ultrafast all-optical switching has only been theoretically reported for bR doped gold nano particles and cytochrome-c (Roy & Yadav, 2014; Yadav & Roy, 2016).

Various attempts have been made and are still in progress to build an all-optical computing system which can meet future demand of high bandwidth, high-speed data transfer, although many challenges need to be overcome that include, low power, high level of parallelism, high scalability, cascadability, high fan out to drive other input stages, logic level restoration, isolation among input and output, absence of critical biasing, complete logic functionality, integration of components to implement and miniaturization using different NLO materials (Avilés-Félix et al., 2020; Banerjee et al., 2020; Ignatyeva et al., 2019; Kuttruff et al., 2020; Lalieu et al., 2019; Melchert et al., 2020; Miller, 2010; Nicholls et al., 2019; Rutckaia & Schilling, 2020; Sasikala & Chitra, 2018).

Challenges

The key challenge in information processing is to achieve ultrafast all-optical switching and ultrahigh bandwidth. SA characteristics of NLO materials have been utilized for designing OR, AND logic gates, whereas RSA characteristics have been used for designing NOT, NAND and NOR logic gates. Major challenge includes for the material which are not only limit to show SA or RSA but can have transition from RSA to SA or vice versa for designing logic gates with the same material under same conditions. Therefore, New materials have to be either discovered, designed or the properties of the existing ones need to be tailored to meet desired specification for practical implementation. Ultrafast optical characterization of materials has to be performed and their photonic response optimized for specific device implementation.

Although ultrafast experimental studies of various organic molecules have been reported in the literature, a detailed investigation essential for realizing all-optical computing needs to be undertaken. On the other hand, theoretical designs of all-optical computing devices have been reported but require

experimental realization. To the best of our knowledge, there are very few materials which have been studied from both experimental and theoretical perspective using femtosecond transient absorption spectroscopy and utilizing their properties for implementing circuitry for all-optical computing.

Motivation

Materials which exhibit both RSA and SA and those materials which exhibit transition from SA to RSA or transition from RSA to SA are of high significance for all-optical switching and designing all-optical logic gates. Theoretical work has been reported for some materials to design all-optical logic gates. Existing studies either show experimental NLO properties and nonlinear dynamics or theoretical implementation of all-optical logic gates for materials.

Therefore, efforts should be made to study femtosecond NLO responses in various organic materials using femtosecond transient absorption spectroscopy, for ultrafast kinetic and spectral changes, which can provide the absorbance, optical density, excited state lifetimes and complete insights into the ultrafast dynamics of the molecules and optimizing the switching characteristics for low power, high contrast and fast switching time, by varying factors that includes intensity, concentration, sample thickness, temperature and wavelength. To the best of our knowledge, a detailed study that encompasses both theory and experiment for ultrafast dynamics of organic materials for optical computing has not been reported. Therefore, the motivation of the proposed study is to undertake a detailed experimental and theoretical analysis of the characterization of various prospective organic materials, using ultrafast tunable laser flash photolysis and use it to realize all-optical computing devices.

Objectives

The objective of the research work is to investigate ultrafast dynamics and switching in the prospective organic molecules, optimize the nonlinear response and design all-optical logic gates. The specific objectives of the study that have been outlined are as follows:

1. To conduct a detailed experimental study of ultrafast optical characterization of organic molecules, that include the following:

   (i) Phthalocyanines, ex. Copper-phthalocyanines

   (ii) Graphene Oxide

   (iii) Ruthenium dioxolene

(iv) Natural photoreceptor proteins, ex. Bacteriorhodopsin and pharaonis phoborhodopsin proteins

2. To analyze the effect of the following parameters on the all-optical switching characteristics, based on the pump-probe configuration:

(i) Pump pulse width and intensity

(ii) Concentration, pH value, temperature and sample thickness

(iii) Pulse frequency

(iv) Absorption cross-sections and life times of excited states

3. To optimize the ultrafast response to realize all-optical ultrafast logic gates.

4. To compare the performance of various organic molecules studied and their device applications.

Methodology

The proposed methodology of the research work is based on the following steps:

Characterization of ultrafast properties of materials using tunable femtosecond laser flash photolysis

Flash photolysis enables measurement of transient absorption in 'spectral mode' and 'kinetic mode'. The sample is photoexcited (pumped) from the ground state to the excited state by a strong excitation (pump) pulse and is brought out of thermal equilibrium. The excited state decays to other intermediate states with a characteristic decay time. By measuring the absorption of the weaker probe pulse which is delayed relative to the excitation pulse, the time evolution of the state under study (ground state, excited state, radical pair, etc.) can be monitored. This so-called transient contains clues to both structural information and dynamics.

The change in optical density, ΔOD, which is typically the difference between pump with probe and "probe-only" spectra, can then be analysed using exponential least-squares fitting algorithms, resulting in transient lifetimes or rate constants.

A detailed study of various materials using ultrafast transient absorption spectroscopy will result in the spectral and kinetic characterization of materials with respect to optical density, absorption coefficient and lifetimes of excited states.

Characterization of nonlinear optical properties

It is proposed to study the nonlinear optical properties of organic materials focusing on nonlinear absorption and nonlinear refraction, which involves high saturable and reverse saturable absorption using z –scan technique.

Characterization of nonlinear material

It is also proposed to carry out further characterization of the organic materials which involves the following:

- ***Tailored Materials for enhanced properties***

Once the analysis on pure organic materials is completed, steps will be taken to enhance their properties for optimal ultrafast optical characteristics. This will be carried out by tailoring materials by doping them with nanoparticles, metal ions, and thin film preparation.

- ***Realization of All-optical computing devices***

It is proposed to utilise the ultrafast dynamics in implementing all-optical logic circuits by experimentally and theoretically analysing various ultrafast boolean and reversible logic gates, and cascading them to realize higher all-optical computing circuits.

Projected Outcomes

The detailed experimental and theoretical study proposed will not only provide important insights into the characterization of the ultrafast response of prospective organic materials, but also its optimization to practically realize ultrafast all-optical computing devices and circuits. The proposed research work would be a significant contribution in the challenging area of all-optical information processing.

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